



# Concatenated logic functions using nanofluidic diodes with all-electrical inputs and outputs

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## ABSTRACT

Nanopore-based logical schemes in ionic solutions typically involve single gates and chemical inputs. The design of computer-like functions requires the consecutive concatenation of several gates and the use of electrical potentials and currents to facilitate the downstream transfer of electrochemical information. We have demonstrated the robust operation of concatenated logic functions using biomimetic nanofluidic diodes based on single pore membranes. To this end, we have implemented first the logic functions AND and OR with combinations of single nanopores using all-electrical input and output signals. The concatenation of these gates allows the output of the OR gate to act as one of the inputs of the AND gate, giving an Enabled-OR logic function. Also, the operation of the OR gate connected with a solid-state transistor, working as a signal inverter, gives a NOR gate. These hybrid electrochemical circuits allow a variety of real time logic functions because of the robust electrical coupling between ionic solutions and electronic elements.

## 1. Introduction

Operational procedures with small electrochemical devices such as energy conversion modules, sensors and actuators, and controlled release drug-delivery dispensers require appropriate responses (outputs) to well-defined stimuli (inputs). This characteristic can be fulfilled by implementing logical schemes on micro and nanoscale devices such as self-assembled monolayers [1,2], nanoparticle arrays [3,4], and biomolecular systems [5]. Alternatively, micro and nanofluidic devices operating in ionic solutions permit logic functionalities by tuning the interaction between the molecules functionalized on the pore surface and free ions [6–15]. In particular, nanopore diodes inserted in polymeric membranes are responsive to chemical, electrical, thermal, and optical input signals, a crucial characteristic for sensors, energy conversion, and signal processing at bioelectrical interfaces [11,16–23]. While the electronic technology dominates logic circuits, demonstrating electrochemically-based logical nanodevices allows information processing in the chemical and biological systems of biomedicine and biotechnology where soft matter micro and nanostructures operating in ionic aqueous environments are commonplace.

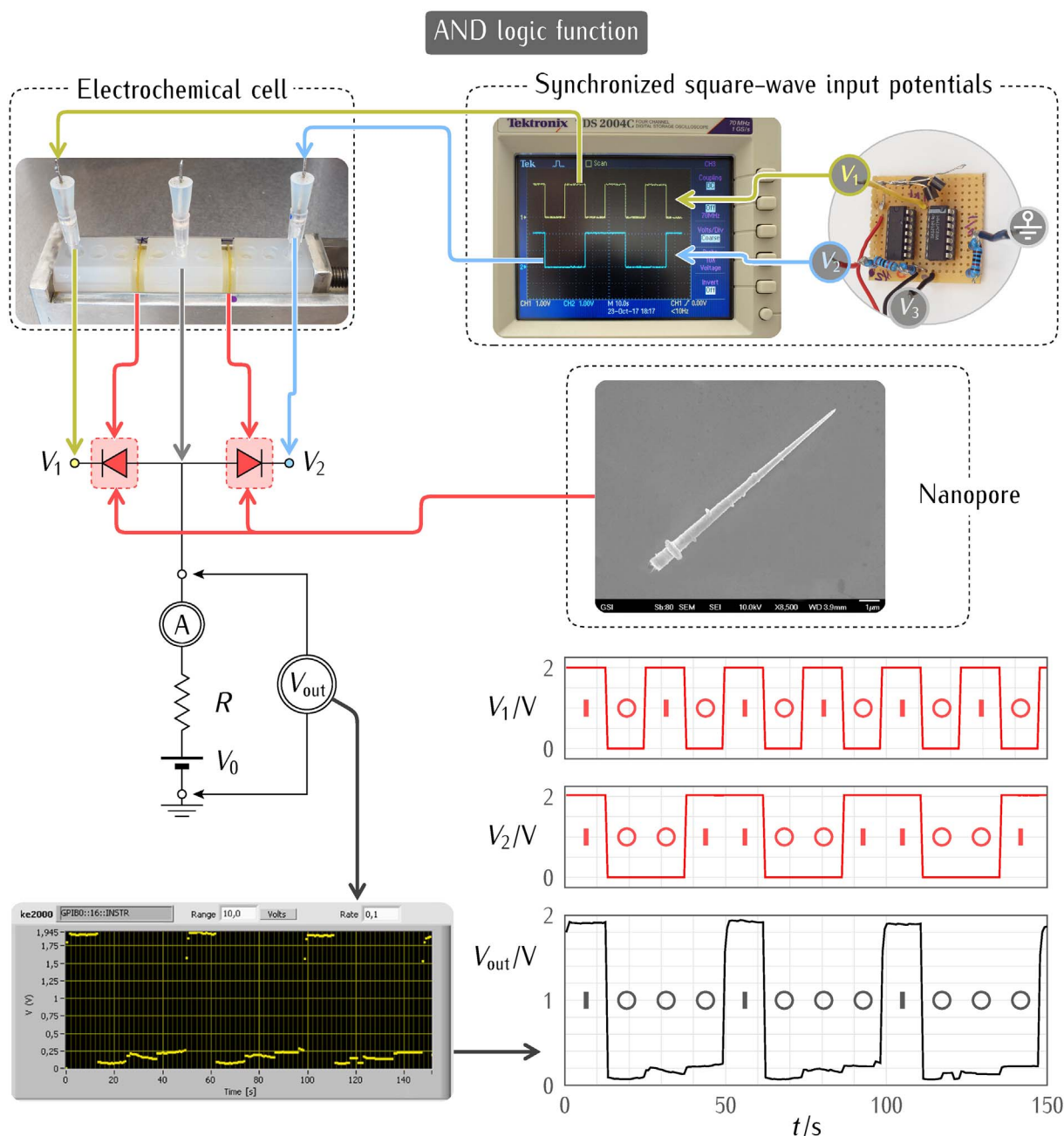
The charged inner walls of nanopores permit a variety of

biomimetic responses in electrical rectification and switching processes [12,17]. Also, the possibility to interconnect these nanostructures with solid-state components and their compatibility with physiological fluids [19,24–28] can be exploited in devices allowing interactive communication with the human body. Most micro and nanopore-based logical schemes previously developed involve single gates and chemical (e.g., solution pH and analyte concentration) inputs [6,7,9,11–16]. However, sensing and actuating with combined chemical/electrical signals may require the design of logical functions of increased complexity for the downstream transfer of information in nanofluidic circuits. Once the first electrical output is acquired from the relevant chemical, thermal or optical input, signal transfer and processing can be implemented by concatenating a sequence of electrical signals in hybrid schemes combining nanofluidic and solid state circuitry. Indeed, the use of electrical signals such as potentials and currents should facilitate the sequential transfer of electrochemical information.

We consider here the design of concatenated logic functions using nanofluidic diodes with all-electrical input and output signals. To this end, we demonstrate first the logic functions OR and AND using combinations of single diodes. The concatenated operation of these gates, assembled in such a way that the output of the first gate constitutes one

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**Fig. 1.** The logic function AND is achieved using nanofluidic diodes obtained by track-etching techniques. Typical gold replicas [27] are shown in the central image. The conical nanopores in single pore membranes are placed in an electrochemical cell and connected to a resistance  $R = 500 \text{ M}\Omega$  at potential  $V_0 = 1.5 \text{ V}$ . The two square-wave input potentials  $V_1$  and  $V_2$  have different periodicity. The input potential  $V_3$  allows the concatenation of this gate with other gates. The ground connection is also shown. A DC 3 V voltage is introduced in the circuit by means the long red and black wires. The potential  $V_{\text{out}}$  is the output signal. The logics '0' and '1' correspond to low and high values of the input and output potentials, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of the inputs of the second gate, allows an Enabled-OR function. Finally, we show that the operation of the OR gate connected with a solid-state transistor working as a signal inverter gives a universal NOR gate. The immobilization of the biomimetic nanopores on solid supports such as polymeric membranes should facilitate the sensing, switching, and re-setting functions in different ionic solution environments.

## 2. Experimental section

### 2.1. Nanofluidic diode

The membrane samples containing single nanopores were obtained from stacks of 12.5- $\mu\text{m}$  thick polyimide (PI) foils (Kapton50 HN,

DuPont) irradiated with swift heavy ions (Au) of energy 11.4 MeV per nucleon at the linear accelerator UNILAC (GSI, Darmstadt). In order to achieve single-ion irradiation, a metal mask with a 200 mm-diameter centered aperture was placed in front of each stack. The ion beam was blocked immediately after a single ion passed through the foil stack and was registered by a particle detector placed behind the samples. The membrane tracks were converted into approximately conical pores by means of asymmetric track-etching techniques [29,30]. SEM images of the nanopore fracture and gold replicas (see Fig. 1 for a typical image) of the conical pores can be found elsewhere [27]. Typical pore radii were in the range 10–40 nm for the cone tip and 300–600 nm for the cone base [27]. Because of the track-etching processes, carboxylate residues were obtained on the pore surface. These residues can be in

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